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CHEMICAL EXCHANGE AS A VERSATILE ISOTOPE SEPARATION PROCESS*

By G. H. Clewett

The successful large-scale separation of the uranium isotopes by two entirely different methods has led to an acceptance of isotope separation as an industrial process. The highly successful gaseous diffusion process has taken its place as a routine operation. At the same time the electromagnetic process has been demonstrating its versatility by supplying separated stable isotopes of nearly all existing kinds for research purposes. Various studies have shown that certain separated stable isotopes have properties of unusual value in nuclear science and technology. It is not unreasonable to believe that a day may come when separated isotopes of many elements will be available in bulk quantities for use in reactor construction and associated projects as well as for laboratory research. Thus isotope-separation processes may be destined to play an even greater role in the future development of nuclear energy than they played in the past. To bring about such a state, it will certainly be necessary to be concerned with costs and to develop those isotope-separation processes which are most efficient.

For the lighter elements, the chemical-exchange method introduced and developed by Urey is generally accepted as being one of the most efficient. The close approach to thermodynamic equilibrium achieved in chemical-exchange processes with resultant minimal expenditure of energy at each stage leads to highly efficient low-cost production.[1] This is true if a suitable isotopic-exchange reaction can be found for the particular element in question. This necessity of searching for suitable reactions for each element prevents the chemical-exchange method from even approaching the versatility of the electromagnetic process. It arises, of course, from the heterogeneity of the chemical properties of the various elements. However, if the chemical and chemical-engineering requirements to be met if a successful process is to emerge are examined and the fields of knowledge which are involved are noted, it will be seen that the progress of research in these fields is such as to make the job continually less difficult. In fact, a condition is approached where it is possible to visualize the chemical-exchange process as having a high degree of versatility. In order to illustrate this, the following criteria have been reviewed.

The specifications to be met by a suitable chemical-exchange system can be grouped into three chemical requirements and three engineering requirements. Reference to specific features of one of Urey's very well known and highly successful separation processes as these six points are listed will serve to clarify each in turn. The enrichment of N^{15} by exchange between NH_3 and NH_4^+ provides an excellent example for this purpose.[2]

CHEMICAL CRITERIA FOR A SUCCESSFUL PLANT

The three chemical criteria may be expressed as follows:

1. The element must be distributed between two phases

$$NH_3\ (gas) - NH_4^+\ (liquid)$$

*Work performed under Contract No. W-7405-eng-26.

2. Rapid isotopic exchange must take place between the chemical forms in the two phases

$$N^{14}H_3 + N^{15}H_4^+ \rightleftharpoons N^{15}H_3 + N^{14}H_4^+$$

3. The equilibrium constant of the isotopic exchange reaction must be other than unity

$$k = \frac{\left[N^{15}H_3\right]\left[N^{14}H_4^+\right]}{\left[N^{14}H_3\right]\left[N^{15}H_4^+\right]} = 1.035 \text{ at } 298°K$$

The reason for the stipulation of the two phases is rather obvious, but the rule can be broken. Thus, at least one successful example of a complete single-phase system is known. Bernstein and Taylor[3] were able to obtain enrichment of carbon 13 by use of the single-phase exchange

$$C^{13}O + C^{12}O_2 \rightleftharpoons C^{12}O + C^{13}O_2$$

These workers were able to move the two species countercurrently by the use of a thermal-diffusion column. Since this is not a very efficient means of obtaining countercurrent flow, it is apparent that a two-phase system is preferable wherein simple pumps can be used. Immiscible pairs which might be used are shown in Table 1.

Table 1

1. Gas-liquid	4. Liquid-solid
2. Gas-solid	5. Solid-solid
3. Liquid-liquid	

Almost all the reported successful chemical-separation processes have consisted of gas-liquid systems, and this type appears to be superior to all others. Examples of these are shown in Table 2.

Table 2

Product	Gas phase	Liquid phase (aq. compds)	Reference
C^{13}	HCN	CN^-	4, 5
C^{13}	CO_2	HCO_3^-	6
N^{15}	NH_3	NH_4^+	2
S^{34}	SO_2	HSO_3^-	7, 8

No clear-cut demonstration of a gas-solid system has been reported, although preferential adsorption of heavy-water vapor on carbon has been proposed as an isotope-separation method.[9] Something of this type might be possible, using fluidization techniques, if the solid particles could be made small enough for rapid isotopic exchange to take place.

There appears to be no reason why a liquid-liquid system should not be satisfactory, yet only one example has appeared — the enrichment of ^6Li by exchange between lithium amalgam and an organic solution of a lithium salt.[10]

Semisuccessful liquid-solid systems have been reported, such as zeolite and ion-exchange resin separations of the isotopes of potassium and lithium.[11] However, these were all inherently batch processes and are not amenable to development into continuous processes.

Solid-solid systems are completely unlikely.

The best possibilities are the two pairs, gas-liquid and liquid-liquid systems. Certainly, wherever possible, it would be desirable to have a gas-liquid system. Generally, however, only the nonmetallic elements provide suitable gaseous species, which leaves the necessity of considering a liquid-liquid

system in most cases. It would appear then that the liquid-liquid type might be regarded as the most versatile.

At this point it may be worth while to examine the third criterion, i.e., that the equilibrium constant for the isotopic-exchange reaction must be other than unity. Urey[12,13] and others[14] have shown how these constants may be calculated, and some degree of qualitative familiarity with the method is quite helpful in understanding which chemical equilibria lead to the largest separation factors. Bigeleisen and Mayer[14] devised two relatively simple formulas for calculating equilibrium constants. They point out several simplifying cancellations which take place in calculating ratios of partition functions for isotopic equilibria. Since the translational and rotational partition functions are classical at room temperature in all cases except for hydrogen, the concern is solely with the vibrational partition function. The most general of the two formulas mentioned above is*

$$\frac{S}{S'} f = 1 + \sum_i G(\nu_i)\, \Delta\nu_i$$

where

$$G = \frac{1}{2} - \frac{1}{\nu} + \frac{1}{e^{\nu}-1}$$

$$\nu = \frac{h\nu}{kT}$$

$\Delta\nu$ = change in ν upon isotopic substitution.

The second formula is for polyatomic symmetrical molecules containing isotopes of a heavy element.

$$\frac{S}{S'} f = 1 + \frac{\Delta M}{24}\frac{m}{M^2} U_1^2 n$$

where $U_1 = \frac{hV_1}{kT}$ and V_1 is the totally symmetric, or "breathing frequency"

ΔM = difference in mass between the isotopes of the heavy element

m = atomic weight of atoms bonded to the central heavy element

M = mass of heavy element

n = number of atoms bonded to the central heavy element

Now, let us examine the ratio of partition functions for some isotopic chlorine molecules.

Mol. ratio:	$\left[\frac{Cl_2^{37}}{Cl_2^{35}}\right]^{1/2}$	$\frac{Cl^{37}O_2}{Cl^{35}O_2}$	$\frac{Cl^{37}O_3^-}{Cl^{35}O_3^-}$	$\frac{Cl^{37}O_4^-}{Cl^{35}O_4^-}$
$\frac{Q_2}{Q_1}$ =	1.0074	1.0313	1.0478	1.0847

*Bigeleisen and Mayer carry the simplification further and obtain

$$\frac{S}{S'} f = 1 + \sum_i \frac{\Delta(U_i)^2}{24}$$

as a very rough approximation.

Thus the equilibrium constants for the following isotopic-exchange reactions would be

$$1/2\ Cl_2^{37} + Cl^{35}O_2 \rightleftharpoons 1/2\ Cl_2^{35} + Cl^{37}O_2 \qquad K = \frac{1.0313}{1.0074} = 1.024$$

$$1/2\ Cl_2^{37} + Cl^{35}O_3^- \rightleftharpoons 1/2\ Cl_2^{35} + Cl^{37}O_3^- \qquad K = \frac{1.0478}{1.0074} = 1.040$$

$$1/2\ Cl_2^{37} + Cl^{35}O_4^- \rightleftharpoons 1/2\ Cl_2^{35} + Cl^{37}O_4^- \qquad K = \frac{1.0847}{1.0074} = 1.077$$

Urey[13] has pointed out that these are progressively larger with added oxygens because of the increased number of vibrational degrees of freedom and the approximate constancy of the vibrational frequencies of all the oxygen compounds of chlorine. That this is not always so is demonstrated by the examples chosen by Bigeleisen and Mayer[14]

$$Si^{28}F_4 + Si^{30}F_6^{--} \rightleftharpoons Si^{30}F_4 + Si^{28}F_6^{--} \qquad K = \frac{1.111}{1.109} = 1.002$$

$$Sn^{120}Cl_4 + Sn^{118}Cl_6^- \rightleftharpoons Sn^{118}Cl_4 + Sn^{120}Cl_6^- \qquad K = \frac{1.00256}{1.00281} = 1.00025$$

Here a considerable cancellation effect is observed because the bonds have become weaker in the octahedral ion than in the tetrahedral ion, and the total restoring force is approximately unchanged. Other examples are available, but these allow a qualitative rule to be formulated for choosing suitable isotopic-exchange reactions. In order to obtain a large separation effect, it is evident that the element in question should exist in one phase unbonded or with only a few weak bonds, and in the other phase it should be bonded with as many bonds as possible, and these bonds should be strong.

When the second chemical requirement of rapid isotopic exchange is examined, some degree of incompatibility is found between the various criteria. The species desired in one phase with many strong bonds is not likely to undergo rapid isotopic exchange with the free element, the ion, or a loosely bonded molecule. If the element is a metal, then one of the first choices of a pair of dissimilar forms is the metal ion in aqueous solution in equilibrium with a chelate complex or other complex of the metal in an immiscible organic solvent. Such coordination entities supply the many strong bonds which lead to a large separative effect, but these bonds are usually considered to be covalent, with resultant negligible exchange. However, if the presence or absence of exchange is accepted as a criterion of bond type, then it must be concluded that covalent bonds are not universally present in such coordination entities since there are numerous examples in which very rapid isotopic exchange takes place. The status of chemical knowledge of the structure of coordination compounds has been briefly summarized recently by Fernelius,[15] and his list of exchange reactions involving coordination compounds contains some which are described as very fast.

Since the success or failure of the Szilard-Chalmers method of producing concentrated radioactive species is intimately related to ease of exchange, a study of some compounds which are not amenable to this treatment is helpful. Fernelius[15] lists a few compounds which apparently exhibit exchange, since the Szilard-Chalmers method was unsuccessful with each.

Table 3

Compound	Reference
Magnesium	
Mg^{++} with $Mg(8\text{-hydroxyquinolinate})_2$ (aqueous E + OH)	16, 17
Zinc	
$Zn\ (pyridine)_2\ (OAc)_2$ with Zn acetylacetonate; Zn acetylacetone-ethylenediamine; Zn benzoylacetonate ammoniate; Zn nicotinyl acetonate; $Zn\ (pyridine)_2\ (SCN)_2$	18
Mercury	
CN^- with $Hg(CN)_4^-$ (pH 10)	19
Manganese	
Mn^{++} with $Mn(C_2O_4)_3^{-3}$	20
Iron	
$Fe(CN)_6^{-4}$ with $Fe(CN)_6^{-3}$	21
Nickel	
CN^{--} with $Ni(CN)_4^-$	19
Ni^{++} with Ni (salicylaldehyde)$_3$	22
Ni^{++} with Ni (ethylene diamine)$_2$	22
Ni^{+3} with Ni (salicylaldoximo)$_3$	22
Ni^{+3} with Ni (salicylaldimino)$_3$	22
Platinum	
Br^- with $PtBr_4^{--}$ and $PrBr_6^{--}$	23, 24
Br in cis-$[Pt(NH_3)_2Br_2]$ with $[PtBr_4]^{--}$	23
Br in $[PtBr_6]^{--}$ with $[PtBr_4]^{--}$	23
Copper	
Cu^{++} with Cu (acetylacetonate)$_2$ in $CHCl_3$	16
Cu^{++} with Cu derivations of salicylaldehyde and related compounds	25

Table 4 — Compounds Not Amenable to Szilard-Chalmers Effect

Compound	Reference
Zinc	
Zn (acetylacetonate)$_2$	26
Scandium	
Sc (acetylacetonate)$_3$	27
Vanadium	
V_2O_3 (8-hydroxyquinoline)$_4$; V(cupferron)$_5$	28
Uranium	
UO_2 (benzoylacetonate)$_2$	29
Manganese	
Mn (acetylacetonate)$_3$; Mn (benzoylacetonate)$_2$	30

There seems to be no marked inconsistency between the conclusions from these experiments regarding bond type and the conclusions from other studies such as magnetic susceptibility measurements and experiments which differentiate between the planar configuration of covalent dsp^2 bonds and the close-packed or tetrahedral arrangement of ions. Some apparent anomalies can be explained by a difference in bond type between the solid compound and a solution of the compound. In other cases certain solvents seem to convert part or all of the covalent molecules into complexes of ionic bonds, thus allowing exchange to take place. The point to be noted is that significant advances are being made in systematizing the knowledge of the structure of coordination compounds and, specifically, the conditions required for isotopic exchange.

The application of this widening knowledge of complex compounds to isotopic equilibria in a quantitative way is some distance away as yet. It has been shown that even an approximate calculation of these equilibrium constants requires a knowledge of the vibrational frequencies arising from bonding of the element in question, and in most cases the shift in frequency upon isotopic substitution must also be known. If these frequencies are unknown and estimates are to be made, then it is, of course, very important to choose the proper atomic model. It will be remembered that the aim is to equilibrate some complex compound or ion of the isotopic element with some simple compound or ion. It would appear that the information about vibrational frequencies would not be difficult to obtain for the simple species in any case. However, where the element in question is a metal, the simple species will most likely be the simple positive ion. It appears that the water of hydration of such metal ions profoundly affects the vibrational partition function, and yet there is no measure of the frequencies of vibration of the metal-oxygen bonds in such hydrates, if, indeed, these frequencies have any physical meaning.

It appears that they are completely absent in raman and infrared spectra. Bigeleisen[31] offers the explanation that perhaps the rate of exchange of water molecules or oxygen atoms about such a metal ion is of the same order of magnitude as the vibrational frequencies which might otherwise be observed.

At any rate, it appears that the presence of water of hydration on these simple metal ions introduces an unknown factor in the problem of calculating isotopic equilibria. On the other hand, it may be of some academic interest that an experimental determination of the isotopic equilibrium factor for suitable individual cases provides an indirect method of learning something about the strength of these ion dipole bonds. Suppose, for example, the equilibrium

$$x_A^{++} + y_{AC_2} \qquad y_A^{++} + x_{AC_2}$$

where A represents a divalent metallic ion, AC_2 represents an extractable complex, and the superscripts represent two different isotopes of the element A. Suppose further that all vibrational frequencies are known for AC_2 and that the equilibrium constant for this exchange reaction is known. It can be seen that through the use of the formulas of Bigeleisen and Mayer, the usual calculation can be reversed, and a numerical result can be obtained for a hypothetical symmetrical vibrational frequency for the hydrated ion. The accuracy of such a calculation is entirely unknown.

It is apparent that the position as regards the versatility of quantitative calculations of isotopic-exchange constants is not a strong one. However, the methods of experimentally measuring these constants are straightforward even though rather tedious, so that the chemical-exchange method of separating isotopes is very little less versatile because of this difficulty in calculating the single-stage factor.

From a chemistry standpoint, if a particular system satisfies the three rules, (a) distribution between two phases, (b) rapid isotopic exchange, (c) equilibrium constant other than unity, then it is ready for engineering development.

ENGINEERING CRITERIA FOR A SUCCESSFUL PLANT

Consider the development problems to be met in applying any exchange system. For the successful application of an isotopic-equilibrium reaction, certain conditions must be met which are more of the nature of chemical-engineering principles. There are various ways of expressing these requirements, but for the present purposes they may be presented as follows.

1. The two immiscible phases of the chemical-exchange system must be efficiently contacted as countercurrent streams.

2. The apparatus must have low holdup and high throughput.

3. Means for achieving total reflux at the top and bottom of the plant must be provided.

When the first two points are considered together, the various means available for efficiently contacting two phases are discussed briefly below. This is where the real advantage of having a system which involves one gas and one liquid phase becomes apparent. That is, a simple packed column suitable for distillation provides a means of contacting these two phases efficiently with very short stage height. Furthermore, such distillation columns have been studied extensively in all sizes and pose no great problem in equipment design. However, as noted earlier, very often it will be impossible to devise a system having one gas phase, and probably liquid-liquid systems must be dealt with in most cases. When two liquids are contacted in an ordinary packed column, the stage heights are usually much greater than those observed with gas-liquid systems. That this should be of concern in no small degree will be manifested if a simple mathematical function in plant design is examined, namely, the time to reach equilibrium.

In most distillation processes the single-stage factors are so large that the entire plant reaches a steady state or equilibrium condition almost as soon as it reaches thermal equilibrium. With an isotope-separation plant, the factor is usually so small and the plant is so long in terms of number of stages that there is a considerable time lag in starting up while the total plant production must go into building up the inventory of enriched material throughout the plant before any product may be withdrawn. The time to reach such a steady state is called the "time to reach equilibrium." Since it is dependent upon the rate of production as well as the size of the plant, it provides a simple measure of plant efficiency. The function is of the following form

$$T = \frac{2h}{E^2}\,\phi(No,N)$$

where h = processing time per stage

$E = \alpha - 1$

$\phi(No,N)$ = function of No and N, which are mole ratio of desired isotope to undesired in product and feed, respectively

The product and feed level are usually fixed. The single-stage separation factor is also fixed for any one system, but since it enters as a square term here, it simply points up further the importance of having a large separation factor. The main concern then is with the processing time per stage. If the rate of exchange of the isotopic reaction is very rapid, the processing time per stage becomes a function of the ratio of holdup per stage to throughput or interstage flow. Thus, in engineering an isotope-separation plant it is very important to achieve a short-stage height with high throughput.

These same features are highly desirable in any plant using a liquid-liquid process, although not so critical in most, and continual progress is being made in devising new and improved equipment for this purpose. Besides various types of packed columns, there are spinner columns,[32] centrifugal separators,[33] as well as simple mixer settlers. Most engineers are familiar with these and many other contacting devices. The development of any design which achieves a significant decrease in the stage height or the ratio of holdup to flow in each stage can mean the promotion to complete feasibility of many otherwise borderline or impractical processes.

The final criterion to be met, of achieving reflux at top and bottom of the plant, is not entirely an engineering problem. First of all, it is a chemical problem of converting the element in question from one chemical form to another. Here again, Urey's process for enriching N^{15} provides an excellent example. The nitrogen, present as ammonium ion in the liquid phase, is converted to ammonia gas by treatment with caustic. The ammonia gas then travels back up the column to the top where it can then be reconverted to ammonium ion with acid. The net result of these chemical processes is the neutralization of an acid with a base to produce a salt. Benedict[1] has pointed out that

this net process and the acid and caustic chemicals used correspond to the power costs or energy costs in other types of separation plants. Because of the large reflux ratio, or ratio of interstage flow to product flow required in an isotope-separation plant, large quantities of chemicals are required usually in this reflux operation. Dependent upon the chemistry involved, equipment must be devised for each different process to accomplish this reflux. It must be done quantitatively with a minimum of holdup.

Since the operating principle of each reflux mechanism is so dependent upon the chemistry of the system itself, very few generalizations are possible. Sometimes the chemistry of a particular system is such as to make it difficult if not impossible to attain this satisfactorily. For these cases it is important to note that a means of obviating the need for reflux has been proposed which is based upon the temperature coefficient of the separation factor.[34] The equilibrium constant for an isotopic-exchange reaction usually varies with temperature in some such manner as shown in Fig. 3. By suitable selection of T_1 and T_2, two towers can be used, a hot tower and a cold tower, operating with two different separation factors. Figure 4 shows how these towers are then connected. If the large factor of the cold tower favors the incoming stream at the top, then the smaller factor of the hot tower favors this same stream somewhat less as it emerges from the bottom of the hot tower. Thus, we have a build-up of desirable component between the two towers as shown in this diagram. The hot tower acts somewhat as a reflux mechanism since it prevents the exit from the system of desirable component by means of its smaller separation factor. Since this eliminates the need for large amounts of chemicals in the ordinary type reflux device, it might be concluded that this dual-temperature method would have somewhat of an advantage. However, this ease of reflux is counterbalanced more or less by the fact that a much larger plant must be built and operated because of the much lower net factor using the dual-temperature process which leads to a larger number of stages. Expressed graphically on a McCabe-Thiele diagram, Fig. 5, it is easy to see that many more stages are required.

These then are the various criteria for successful application of chemical exchange to the separation of isotopes. It is believed that the advances now being made in the fields of knowledge mentioned above are such as to materially extend the applicability of this method.

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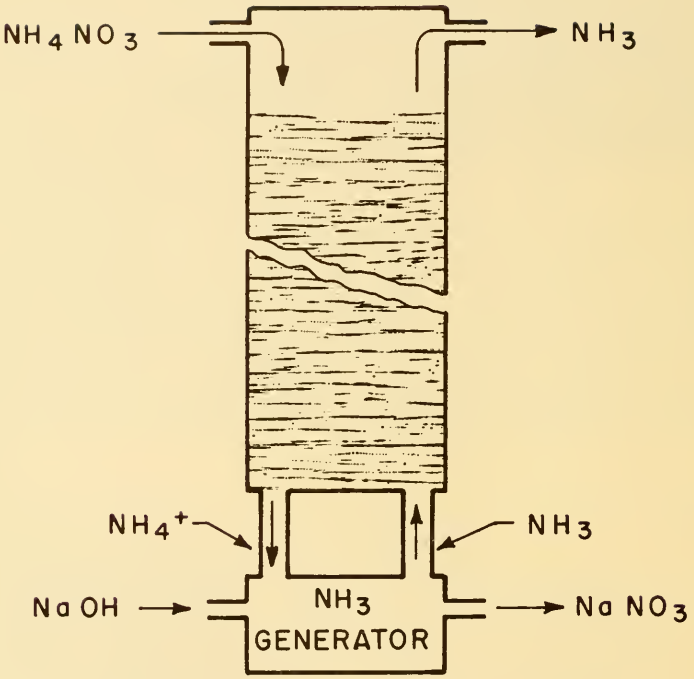

Fig. 1--$N^{15}H_3$ (gas) + $N^{14}H_4^+$ (sol.) $\rightleftharpoons$ $N^{14}H_3$ (gas) + $N^{15}H_4^+$ (sol.) [H. G. Thode and H. C. Urey, J. Chem. Phys., 7: 34 (1939)].

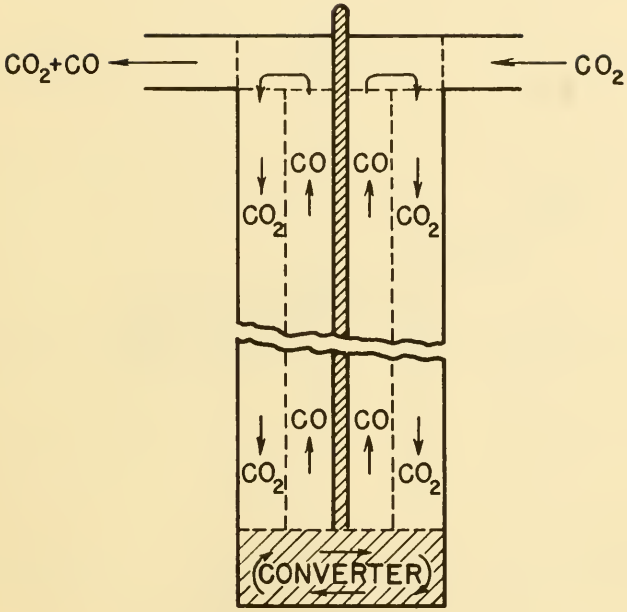

Fig. 2--$C^{12}O_2$ (gas) + $C^{13}O$ (gas) $\rightleftharpoons$ $C^{13}O_2$ (gas) + $C^{12}O$ (gas) [R. B. Bernstein and T. I. Taylor, J. Chem. Phys., 16: 903 (1948)].

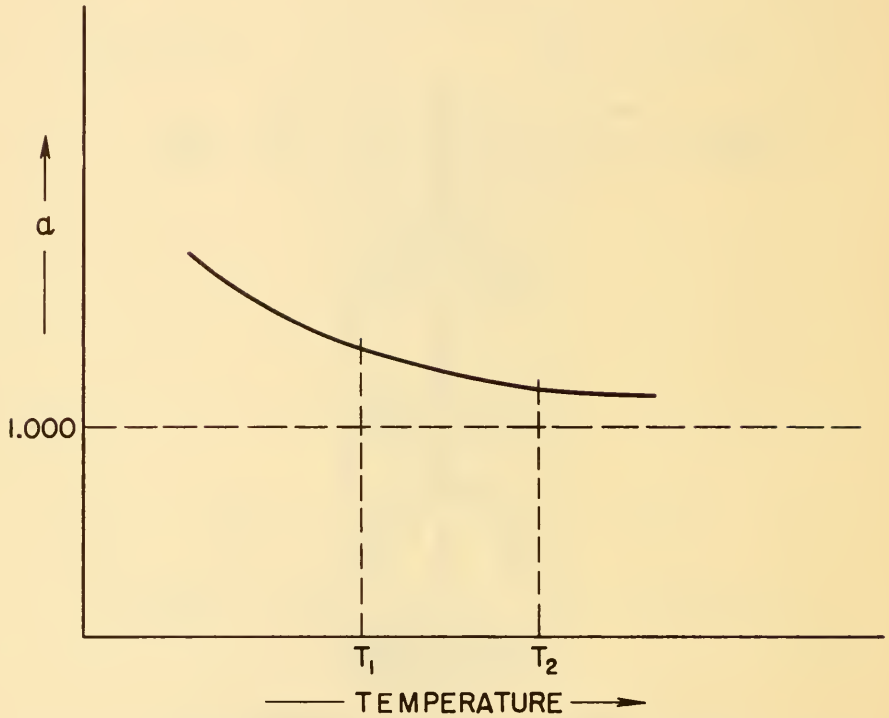

Fig. 3--Variation of the equilibrium constant for an isotopic-exchange reaction with temperature.

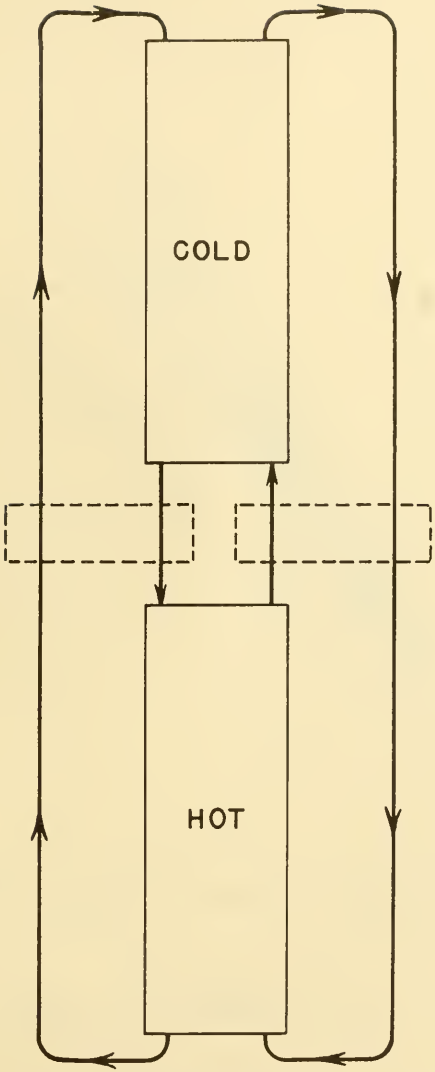

Fig. 4--Diagram of hot and cold towers connected.

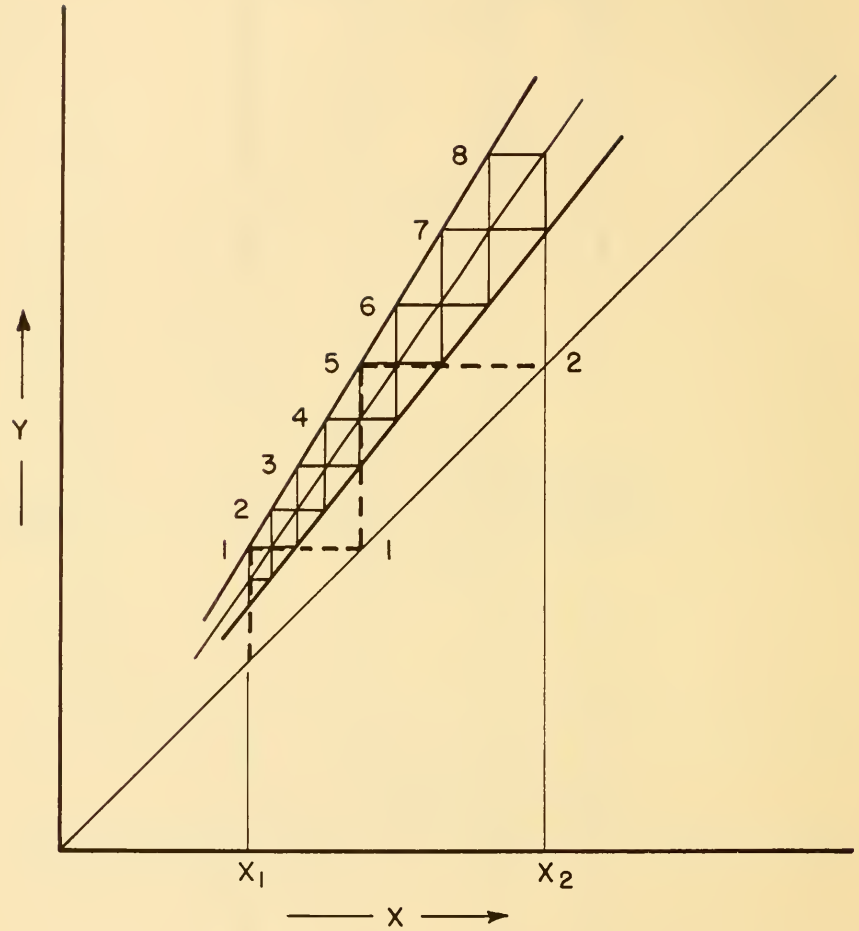

Fig. 5--McCabe-Thiele diagram.

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